Magnetic Anisotropy in $Ca_3Ru_2O_7$

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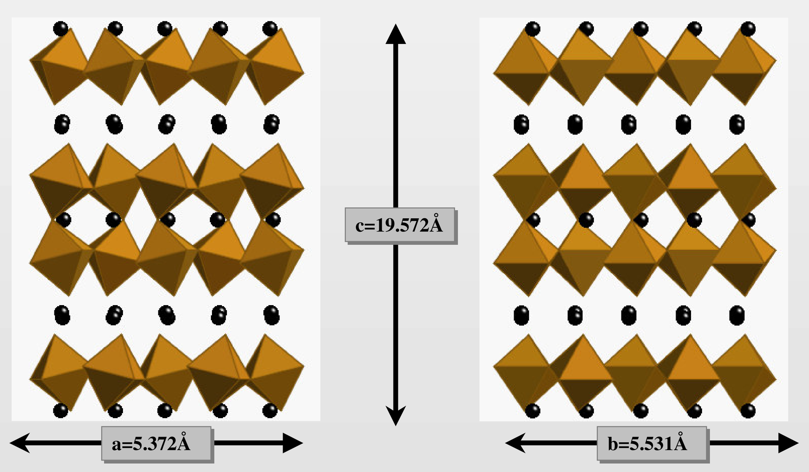
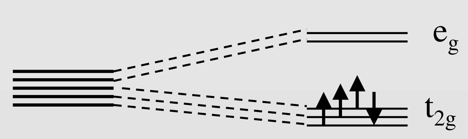
$Ca_3Ru_2O_7$ is a member of the Ruddlesden-Popper Series with the general form: $(Ca,Sr)_{n+1}Ru_nO_{3n+1}$ where n defines the number of RuO_6 planes in the unit cell. To date, members with n = 1,2,3,∞ have been grown, and collectively they exhibit a tremendously rich array of behaviors including the first observed 4d ferromagnet ($SrRuO_3$, T_C=160K), an unconventional superconductor (Sr_2RuO_4 , T_c=1.5K), a Mott metal-insulator transition (Ca_2RuO_4, T_{MI}=357K) and a recent flurry of interest at the suggestion of a quantum critical point in $Sr_3Ru_2O_7$ tuned by an external magnetic field. Substitution of isovalent, but smaller Ca ions for Sr in the bilayered compound distorts the lattice from nearly tetragonal (a=b≠c) to an orthorhombic (a≠b≠c) crystal structure, resulting in a sharp tilting of the RuO_6 octahedra within adjacent planes. This distortion leads to radically different phase diagrams when a magnetic field applied along the different directions to breaks the symmetry. A number of other unusual phenomena are also observed including magnetically induced superthermal effects.



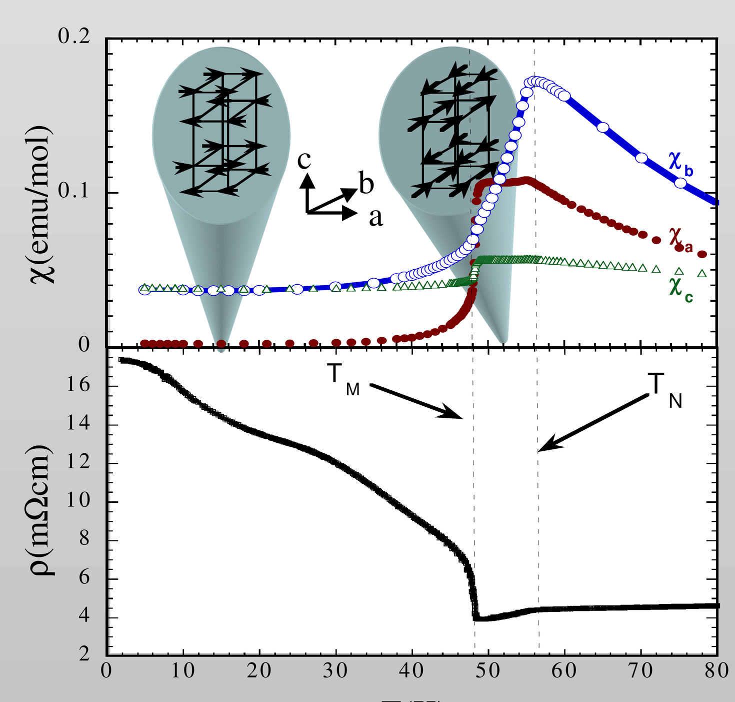
H || a **H || b**

Crystal electric fields of the 6 surrounding oxygens removed degeneracy from the Ru 4d orbitals

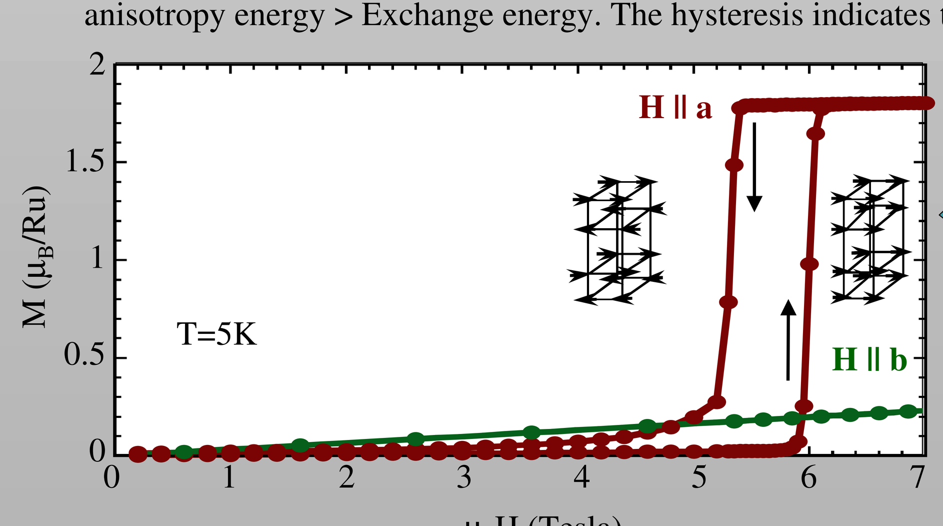


Ru^{4+} $(4d^4)$ S=1

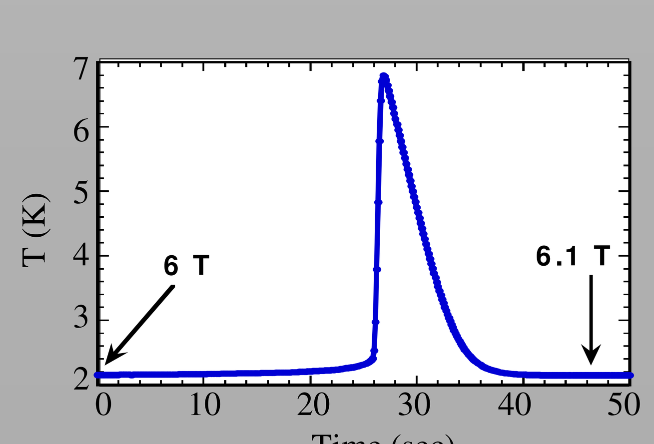
When H = 0, the system orders as an type A antiferromagnet(AF) at T_N=56K, and then undergoes a metal-nonmetal transition at T_M=48K. The resistivity is qualitatively similar along all directions. At T_M the spins also spontaneously reorient changing from a textbook AF to an Ising like state, with a gap in the spin-wave spectrum.



This system undergoes a metamagnetic transition at 6 Tesla when H || a, indicating that the anisotropy energy > Exchange energy. The hysteresis indicates the transition is first order.

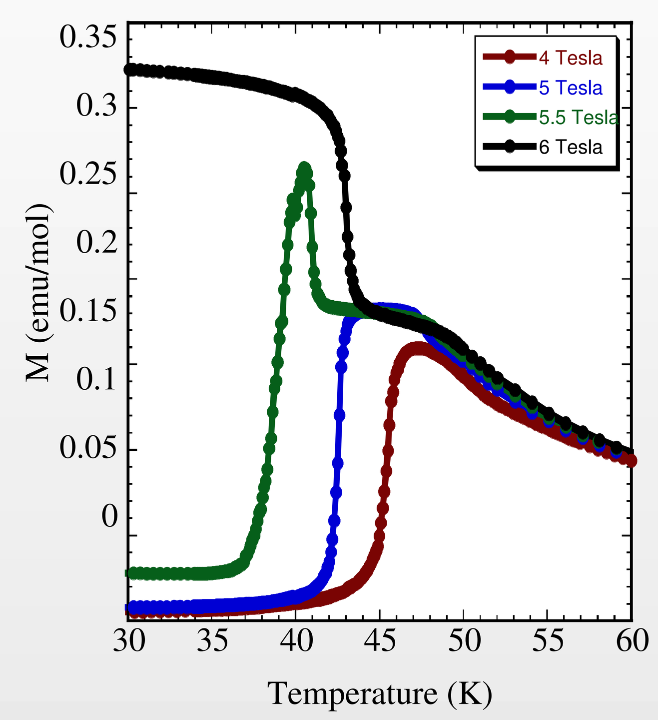


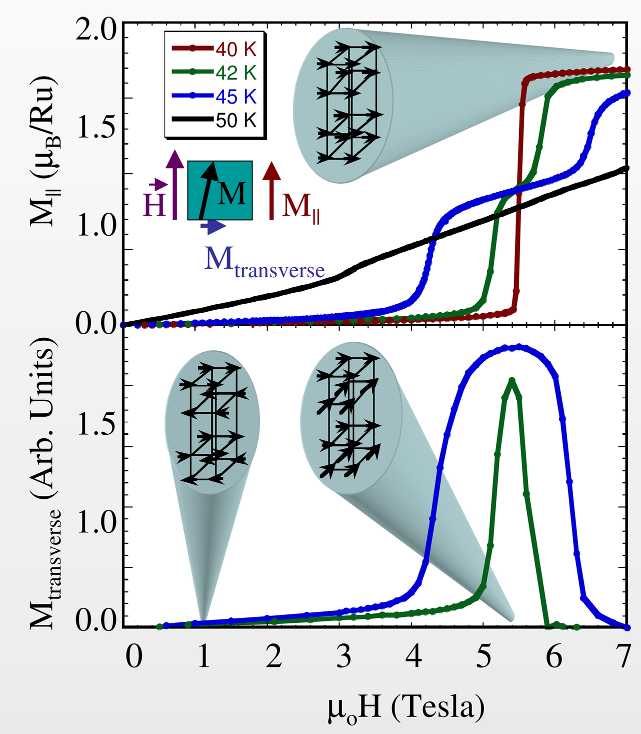
From χ(T=0) and H_c it is possible to obtain the anisotropy and exchange energies.
H_A = 22.4 T
H_E = 15 T
So $H_A > H_E$



Measurement of a 3 mg sample weakly connected to a 2K bath. As the applied magnetic field is swept upwards from 6T, the sample undergoes a metamagnetic transition with spontaneous evolution of heat as the spins flip. The energy released can be calculated, in this case 8.4 ± 0.2 J/mol. A similar transition occurs sweeping the field down at ~5.15T. The total heat released for both directions, 16.8 ± 0.3 J/mol compares favorably to the hysteresis loss: $\oint MdH$ = 17J at T=2K.

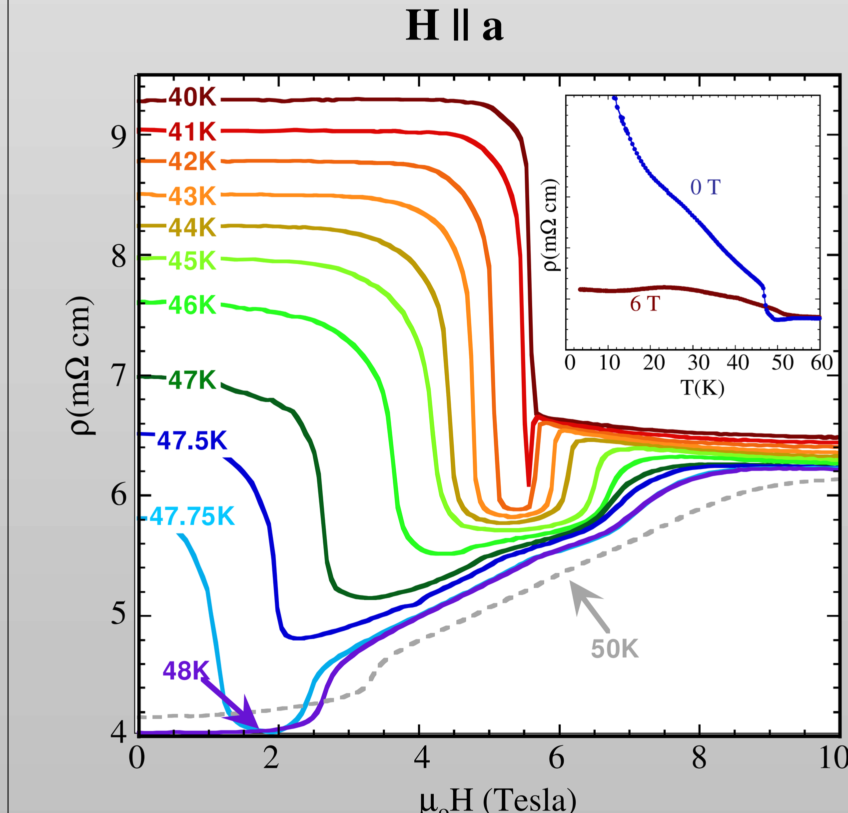
A Magnetic Field applied parallel to the a-axis



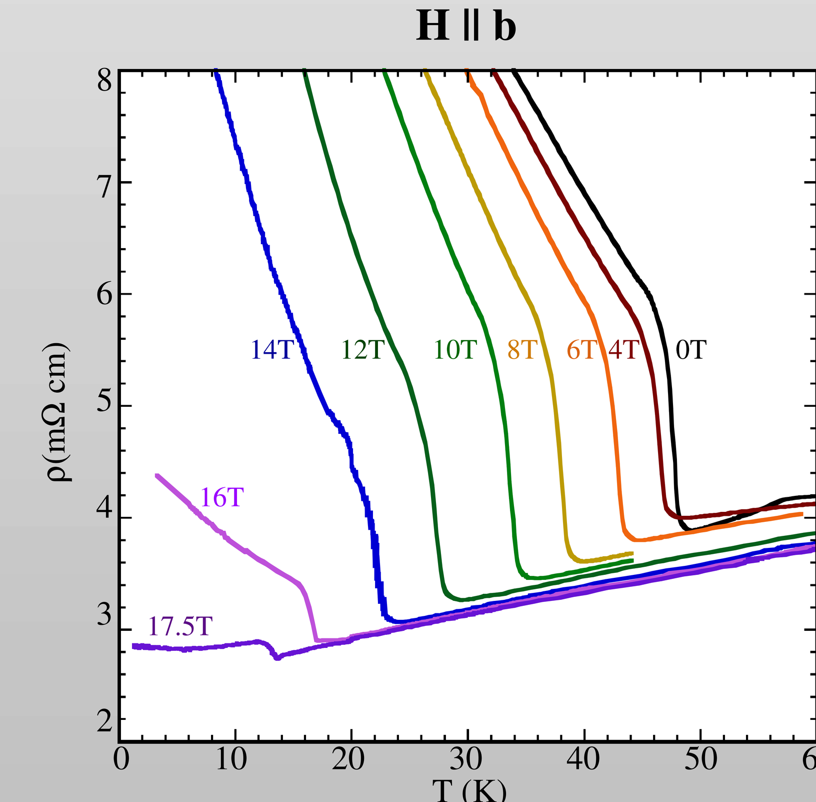


A magnetic field applied parallel to the a-axis leads to a complex phase diagram near T_M(H=0). Some of the effects are shown in the magnetic measurements above, with "spin cartoons" suggesting the magnetic phases. These results are further elucidated by the isothermal magnetoresistivity data, ρ(H||a) shown below on the left, where the inset shows the general behavior for ρ(T) for magnetic fields parallel to the a-axis below 6T (blue) and above 6T (red). By contrast the resistivity, ρ(T,H||b) is shown on the left for different fields with H ||b, where the magnetic field clearly suppresses T_M.

H || a

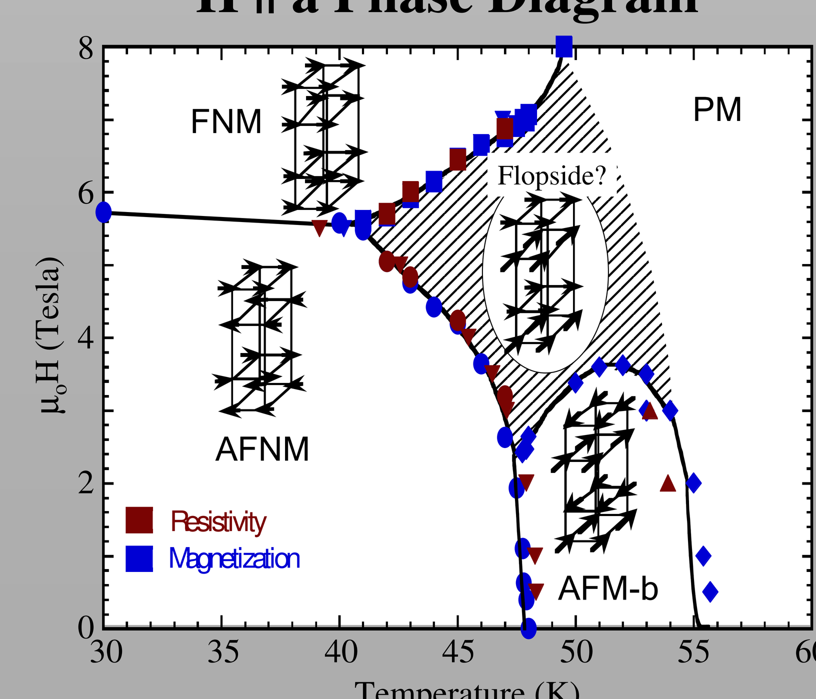


H || b

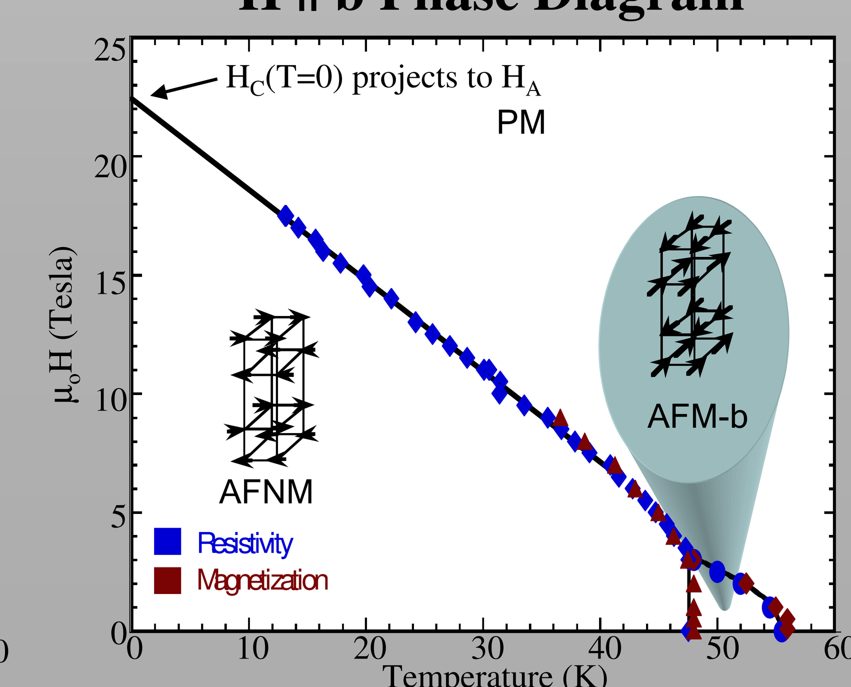


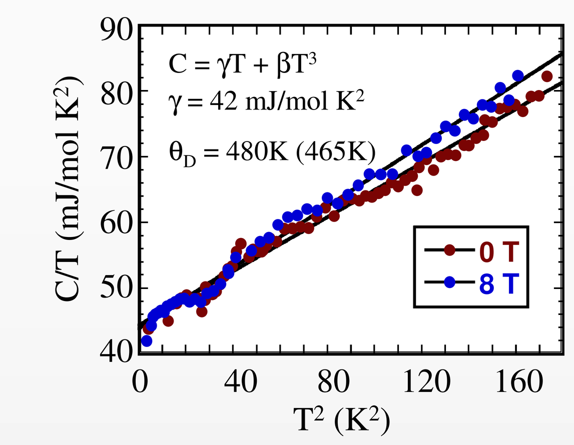
The magnetoresistivity plots above suggest the general form of the phase diagrams shown below. The phase lines were determined from a large number of measurements including: ρ(T), ρ(H), M(T), M(H), and C(T). The phases include: ferromagnetic non-metal(FNM), antiferromagnetic non-metal(AFNM), antiferromagnetic metal(AFM-b) with spins along the b-axis, a Flopside state, and paramagnetic metal (PM) at high temperatures.

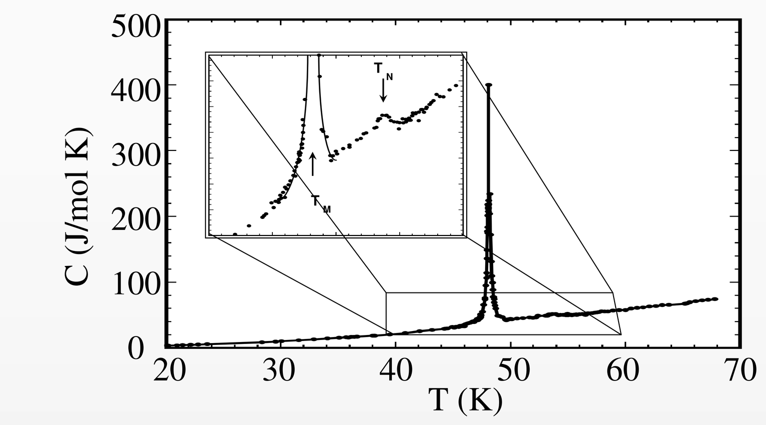
H || a Phase Diagram



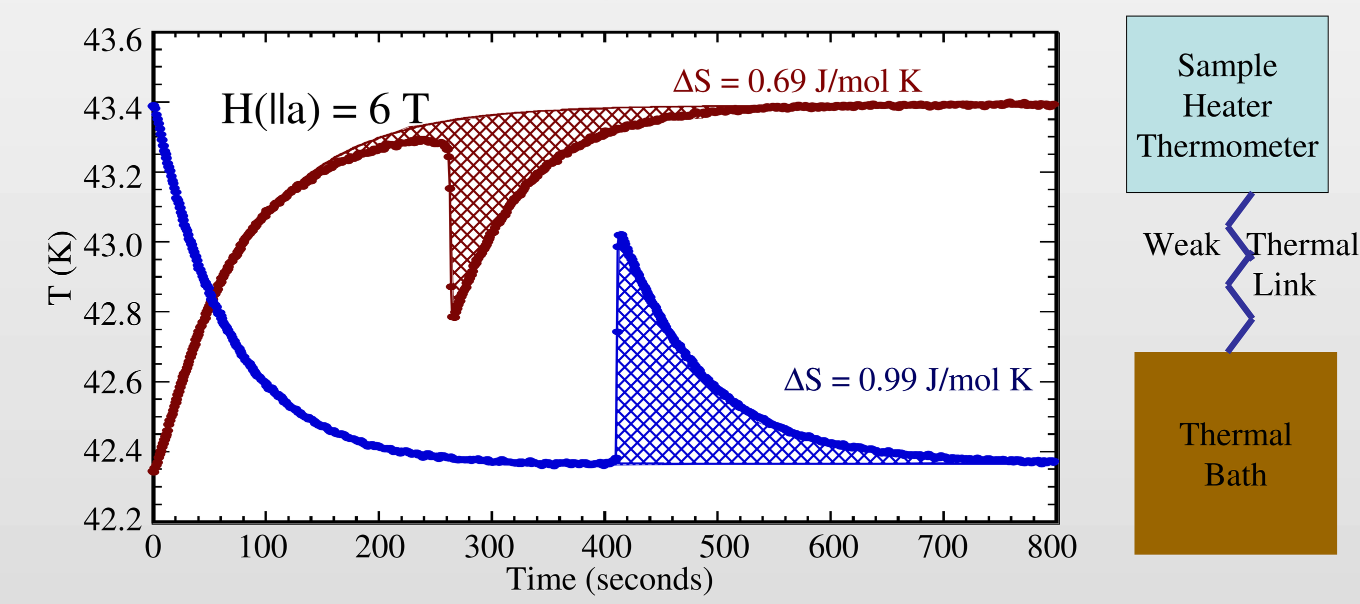
H || b Phase Diagram





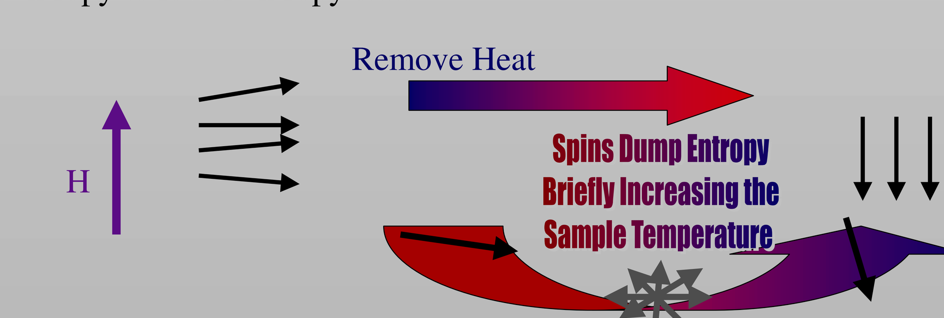


The low temperature electronic contribution to the specific heat is strongly enhanced and shows no change crossing the 6T metamagnetic transition. The sharp peak at T_M (48K) removes significant entropy (2.7 J/mol K) suggesting a first order transition. There is a very weak transition at 54K indicative of the AF ordering, however the entropy removed (~0.15 J/K mol Ru) is only a very small fraction of the expected spin entropy (Rln(2S+1) = 8.3 J/K mol Ru), suggesting the spins are itinerant at T_N.



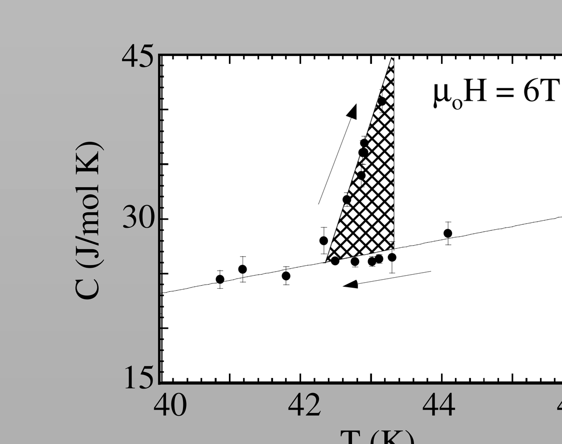
When a magnetic field is applied along the a-axis, the extremely unusual phenomenon of superheating and supercooling has been observed. As shown in the above cartoon, the sample is connected only very weakly to a thermal bath, so that application of a constant amount of power will warm the sample until the heat flowing out of the weak link is equal to the power supplied by the heater. When the heater is turned on at the proper temperature, the sample initially warms, and then spontaneously cools extremely rapidly before warming back up again as the spins steal energy from the lattice to flop to a less ordered state.

After switching the heater off, the sample initially cools, but again, at a critical temperature it suddenly self-heats before cooling back to the bath temperature as the spins dump energy in the lattice to move from a high entropy to lower entropy state.



What's the story with the Entropy?
ΔS = 0.99 J/mol K Cooling
ΔS = 0.69 J/mol K Warming
0.30 J/mol K Missing !

There is a hysteresis in the heat capacity: The difference between the warming and cooling data sets is given by the cross-hatched area: ΔS = 0.28 ± 0.04 J/mol K



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